

Characteristics of a tropospheric ozone profile and implications for the origin of ozone over subtropical China in the spring of 2001

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[1] During the Transport and Chemical Evolution Over the Pacific (TRACE-P) period in the spring of 2001 we launched an ozonesonde at three Chinese sites: Kunming (102.68°E, 25.03°N), Hong Kong (114.17°E, 22.31°N), and Linan (119.75°E, 30.30°N). The sites extend from subtropical southwestern China close to the Southeast (SE) Asian border, to the southeastern Asian coast, and to the edge of the middle latitudes of central eastern China, respectively. The aims of the study are to provide ozonesonde data within the source regions of the Chinese mainland, to investigate the source of tropospheric ozone (O₃), and to investigate to what extent SE Asian biomass burning emissions impact both tropospheric O₃ over the subtropical Chinese mainland and O₃ outflows to the Pacific. The results show that there are substantial variations in vertical O₃ distributions over these sites, with low O₃ values in the upper troposphere of Hong Kong, high O₃ values in the middle and upper troposphere of Linan, and frequent O₃ enhancements in the lower troposphere of Hong Kong and Kunming. The low values in the upper troposphere over Hong Kong in the spring of 2001 were not usually observed from 1993 to 2000 and are the result of the transport of O₃-depleted air from the intertropical convergence zone of equatorial SE Asian regions following the eastern Asia local Hadley circulation. Such transport processes do not affect the higher latitude at the edge of the middle latitude of Linan, where stratospheric O₃ is the major contributing source to middle and upper tropospheric O₃. The O₃ enhancements over the lower troposphere of Kunming and Hong Kong are caused by SE Asian biomass burning emissions. Such enhancements are frequently observed over Hong Kong, less often over Kunming, and scarcely ever over Linan. Our analysis shows that biomass burning emissions from SE Asia in the spring of 2001 mainly affected the southern parts of the subtropical Chinese region. **INDEX TERMS:** 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; **KEYWORDS:** tropospheric ozone, stratospheric intrusion, pollutant transport

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1. Introduction

[2] One of the objectives of the Transport and Chemical Evolution Over the Pacific (TRACE-P) study is to better understand the chemical evolution of tropospheric O₃

associated with Asian outflow over the western Pacific and the processes that control this evolution [Jacob *et al.*, 2003]. During March–April 2001, when the two NASA aircraft were flying over the western Pacific, we performed simultaneous ozonesonde launchings at three locations in southern China. The first aim was to supplement the aircraft measurements by providing ozonesonde data within the source regions of the Asian continent, inside mainland China. The second aim was to investigate the source of tropospheric O₃ and to find the extent of the impact of SE Asian biomass burning emissions on tropospheric O₃ over subtropical China and on O₃ outflow to the Pacific.

[3] The large-scale air pollutant emissions in the Indonesian region of Southeast (SE) Asia and southern Asia as a

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Figure 1. Map showing the ozonesonde launching sites in Kunming, Hong Kong, and Linan.

result of forest fire, biomass, and biofuel burning has drawn international focus recently. This pollution strongly impacts the chemical composition of the atmosphere and the climate system [Fujiwara *et al.*, 1999; Matsueda and Inoue, 1999; Tsutsumi *et al.*, 1999; Chan *et al.*, 2001; Lelieveld *et al.*, 2001; Thompson *et al.*, 2001; United Nations Environment Programme and Centre for Clouds, Chemistry and Climate, 2002]. The effects of biomass burning in the tropical Indonesian and SE Asian regions on tropospheric and surface O_3 in the same regions, especially during El Niño events, have been reported [Pochanart *et al.*, 2001; Yonemura *et al.*, 2002a, 2002b]. The possible impact of biomass burning emissions from the SE Asian subcontinent on tropospheric O_3 over the subtropical Chinese mainland was first suggested by Liu *et al.* [1999], who noted a regular substantial enhancement of tropospheric O_3 in the springtime over the lower troposphere (2.0–4.5 km) of Hong Kong. Chan *et al.* [2000] showed with a case study that the O_3 enhancements are the result of photochemical O_3 produced by biomass burning emissions from the Indo-Burmese region of the SE Asian subcontinent. Chan *et al.* [2003b] revealed through satellite data analysis that the biomass burning activities in the Indo-Burmese and Indian-Nepalese regions reach their full strengths in the dry season, from late February to mid-April, coinciding with the springtime tropospheric O_3 maximum over Hong Kong [Chan *et al.*, 1998]. A chemical composition and transport characteristics analysis of the O_3 -rich air masses reaching Hong Kong showed that these air masses traverse over a large geographical region of the subtropical Chinese mainland before outflowing to the Pacific [Chan *et al.*, 2003b]. These

findings inspire us to explore the possible impacts of biomass burning emissions from SE Asia on the tropospheric O_3 over the general subtropical Chinese regions and on outflows to the Pacific.

[4] Talbot *et al.* [1996] found that the strong cyclonic activities over Asia during the 1994 Pacific Exploratory Mission (PEM)-West A experiment (September–October 1991) could have transported biomass burning emissions from SE Asia or emissions from the extensive use of biomass materials to the upper troposphere. Blake *et al.* [1997] noted that the polluted air masses observed over the western Pacific during the 1994 PEM-West B experiment are rich in biomass burning emission tracers, including hydrocarbon (HC) and halogen HC tracers such as methyl chloride. Folkens *et al.* [1997] showed that deep convection can inject emissions from SE Asian biomass burning to the near-tropical tropopause. Harris *et al.* [1998] pointed out with evidence from the CO, O_3 , and CH_4 relationship that the strong horizontal O_3 gradient from the SE Asian continent to the Mauna Loa Observatory (MLO) in the central Pacific is related to the springtime tropospheric O_3 maximum observed at the MLO station. However, the authors cannot identify the sources contributing to the O_3 gradient. Chan *et al.* [2003b] showed with forward trajectory that the O_3 -rich air masses observed over Hong Kong and outflowing to the western and central Pacific from the subtropical Asian coast can reach the western coast of the United States within 10 days. In this paper we present the ozonesonde measurement data observed at three southern Chinese stations. The objective is to investigate the characteristics of tropospheric O_3 profiles and to explore their origin over the large geographical region of the subtropical Chinese mainland.

2. Experiment

[5] During the TRACE-P period, when the two NASA aircraft were flying over the eastern Asian coast and the western Pacific, an ozonesonde was launched at three Chinese sites at Kunming, Hong Kong, and Linan; these sites cover the region from subtropical southwestern China close to the SE Asian border, to the southeastern Asian coast, and to the edge of middle latitudes in central eastern China, respectively (Figure 1). Table 1 summarizes the characteristics of the launching sites and of the ozonesonde launches at these sites. The Linan launching site (119.75°E, 30.30°N) is primarily a rural area. It is 134 m above sea level (asl). There were a total of 28 launches from 3 March to 13 April 2001, with a trial launch on 21 February. The O_3 profiles were available everyday from 3–13 March. After that, there was at least one profile every other day until 13 April, with the exception of from 4 to 7 April, during

Table 1. Summary of Ozonesonde Launching Sites and Soundings During the TRACE-P Period

Site	Location		Altitude, m asl	Site Characteristics	Number of Soundings	Date of Sounding	
	Longitude, °E	Latitude, °N				Begin Date	End Date
Kunming	102.68	25.03	1820	suburban area on the edge of the Tibetan Plateau	27	1 March 2001	13 April 2001
Hong Kong	114.17	22.31	65	urban area on the southeastern coast of China	69	March 2000	June 2001
Linan	119.75	30.30	134	rural village on the central eastern coast of China	28	3 March 2001	13 April 2001

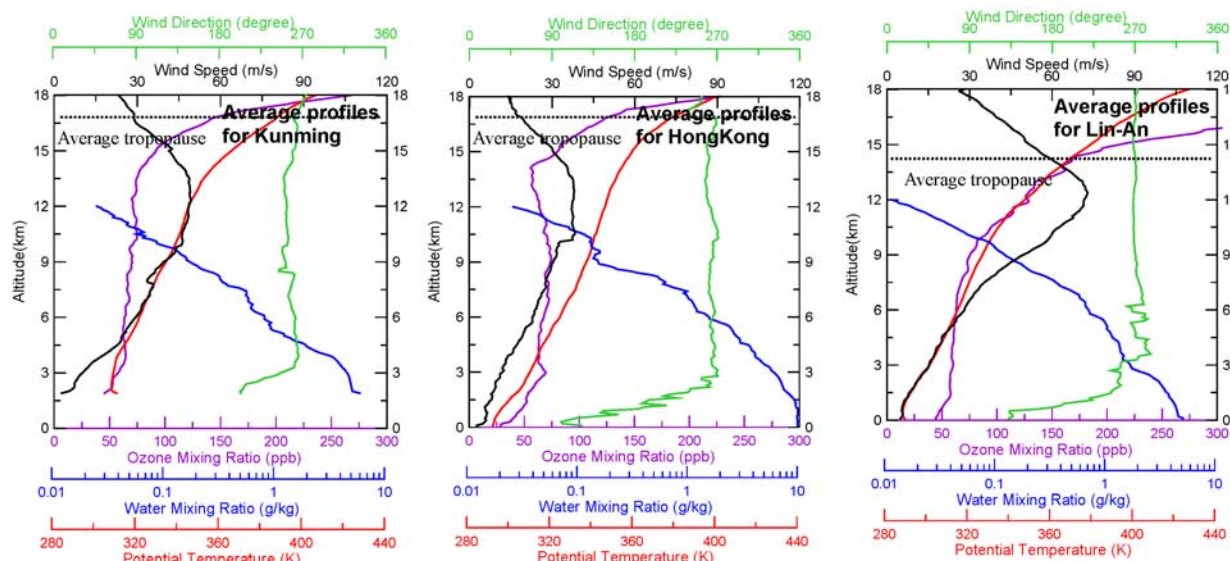


Figure 2. The average profiles of ozone, the water mixing ratio, potential temperature, wind speed, and wind direction over (left) Kunming (28 February to 13 April), (middle) Hong Kong (2 March to 25 April), and (right) Linan (21 February 13 April). The tropopause heights are marked by dotted lines.

which only two profiles were available. The Kunming launching site (102.68°E, 25.03°N) is situated on the edge of the Tibetan Plateau in a suburban area, with an altitude of 1820 m asl. There were a total of 27 launches from 1 March to 13 April, with a trial launch on 28 February 2001. The launching site in Hong Kong is the Upper Air Monitoring Station of the Hong Kong Observatory. A detailed description of the launching site in Hong Kong (114.17°E, 22.31°N) can be found in the work of *Chan et al.* [1998]. The launching in Hong Kong for the TARCE-P mission began in March 2000 and lasted until June 2001, with a frequency of one launch per week and higher frequencies when the two aircraft were flying close to the Hong Kong region. In all three sites the launching was performed at 0500–0600 UT.

[6] Tropospheric ozone profiles were measured by in situ electrochemical concentration cell ozonesondes (Model 6a, Vaisala) that were coupled with a standard radiosonde (RS80-15GE, Vaisala). A Vaisala DigiCora MW 15 receiving system was used. The ozonesonde launching system, the experimental procedures, and the quality control and assurance procedures used were similar to those described by *Chan et al.* [1998, 2003b]. The ozone data measured by ozonesonde have precisions from ± 3 to $\pm 12\%$ in the troposphere. The accuracies for individual ozonesonde soundings are $\pm 6\%$ near the ground and 7–17% in the high troposphere, where ozone mixing ratios are low [*Komhyr et al.*, 1995].

3. Results and Discussion

3.1. Characteristics of Tropospheric Ozone Profiles

[7] Figure 2 shows the average profiles of the O_3 mixing ratio, water mixing ratio, potential temperature, wind speed, and wind direction below 18 km for the three launching sites. The average profiles comprise all the profiles available

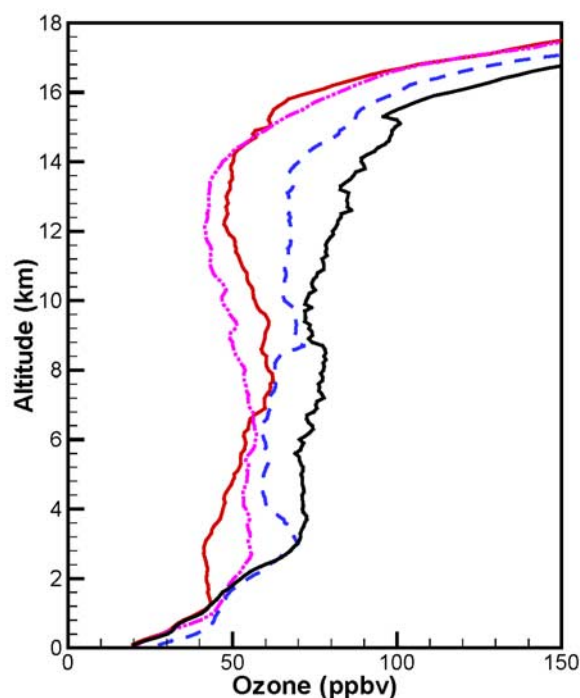


Figure 3. The monthly average ozone profile for December (dash-dotted line), February (solid line), March (dashed line), and April (bold solid line) over Hong Kong from October 1993 to December 2001.

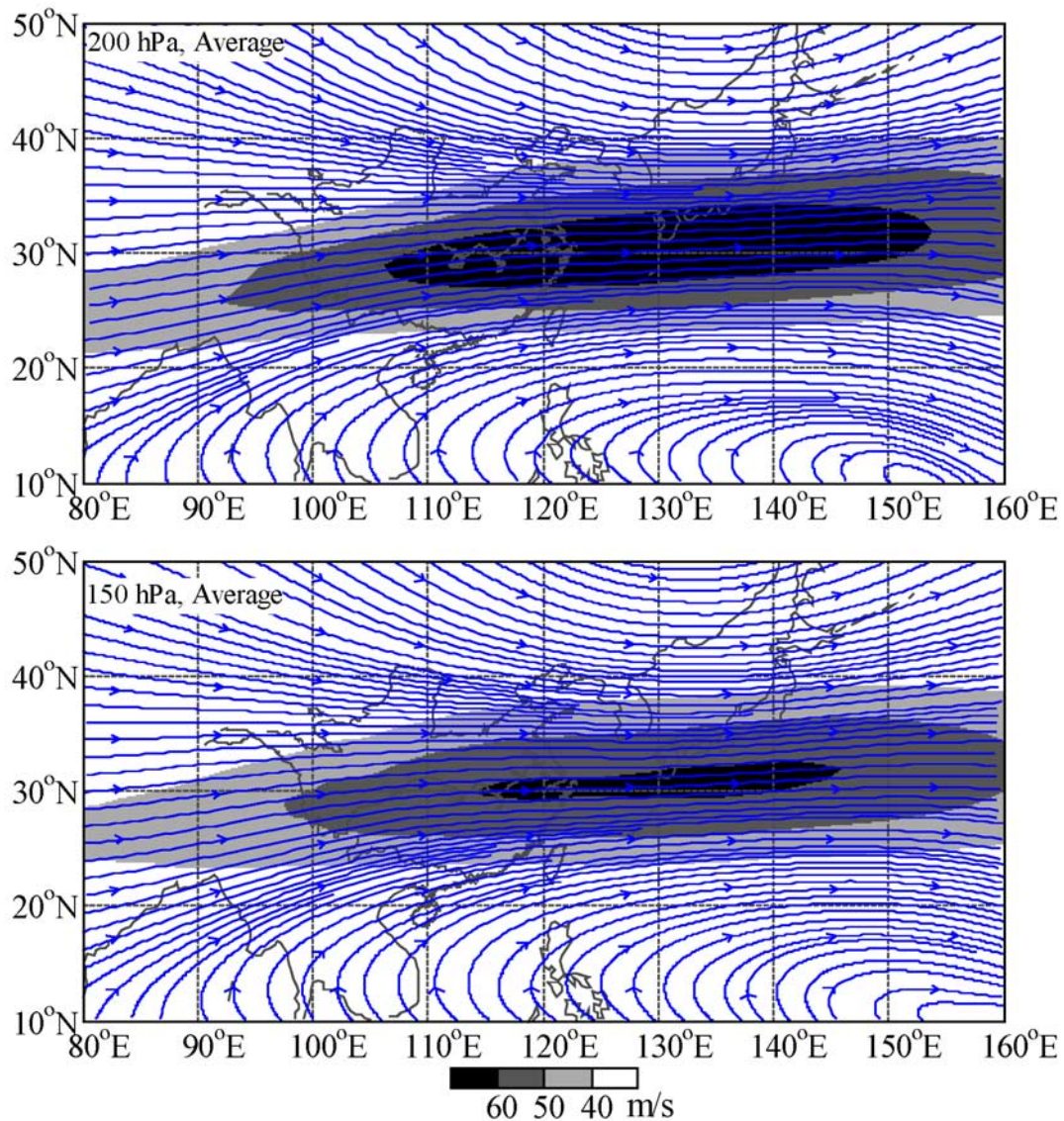


Figure 4. The average streamlines and isotachs (m s^{-1}) at (top) 200 and (bottom) 150 hPa from 1 March to 13 April 2001.

during the TRACE-P period. Average thermal tropopause heights, as determined from the temperature profiles measured by radiosondes, are also marked in Figure 2 (dotted lines). The average thermal tropopause height was 16.9 km (14.6–18.1 km) over Kunming, 16.7 km (14.9–18.1 km) over Hong Kong, and 13.8 km (9.9–18.1 km) over Linan. The average O_3 mixing ratio for these three sites ranged from 50 to 75 ppbv in the free troposphere and from 25 to 60 ppbv in the boundary layer below 1.5 km. Comparing the three O_3 profiles, we noted that there are substantial differences in the distributions of O_3 in the subtropical atmosphere of southern China. At the edge of the middle latitudes over Linan the O_3 mixing ratio showed a steady increase from <50 ppbv close to the surface to ~ 75 ppbv at 9.0 km, from where it increased substantially to >150 ppbv at 13.5 km above ground and to higher values at the higher altitudes. Note that the sharp O_3 increase between the

altitudes of 6.0 and 13.5 km closely followed the increase of potential temperature. The potential temperature increased from ~ 310 K at 6.0 km to 360 K at 13.5 km over Linan, while it increased from ~ 320 K to 345 K at the same altitudes over Kunming and Hong Kong.

[8] In southwestern China, over Kunming, the tropospheric O_3 mixing ratio ranged from 50 to 75 ppbv. Readings showed a steady increase from the surface to ~ 13.5 km, where readings started to increase sharply. The increase from the surface to 13.5 km is slow. The sharp increase from 13.5 km coincided with an increase in the potential temperature, which indicates the tropopause. The O_3 mixing ratio below 13.5 km ranged from 50 to 75 ppbv. We note that there was a distinguishable local O_3 peak, with higher O_3 at 3.0–4.5 km altitude. In southeastern China, over Hong Kong, the O_3 mixing ratio ranged from ~ 25 ppbv at the surface to ~ 50 ppbv at 1.5 km above ground. In the free

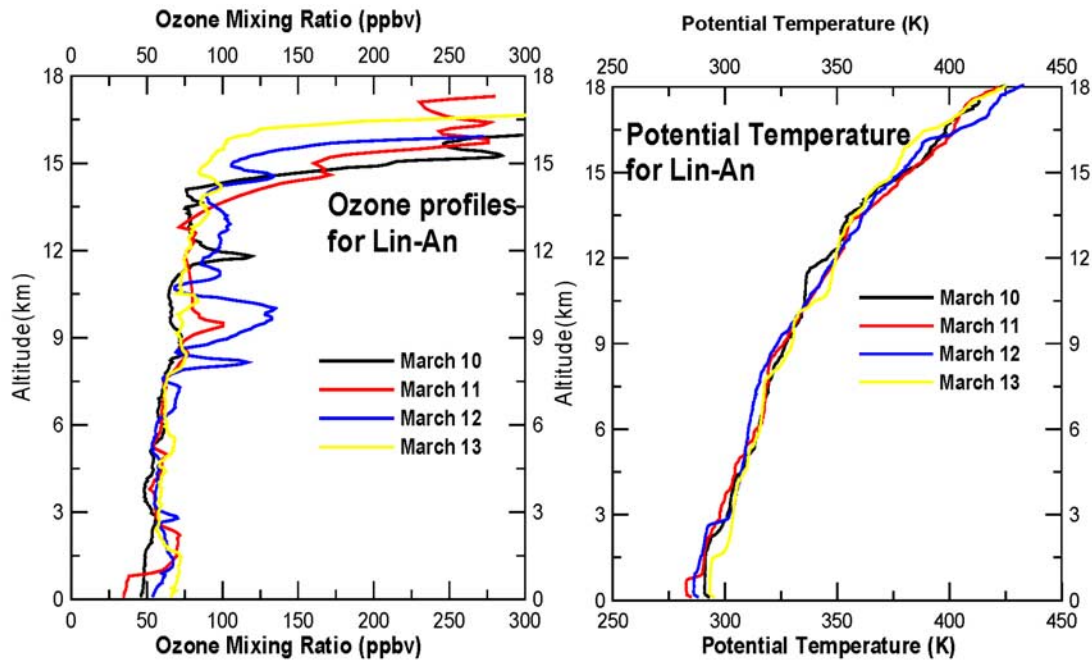


Figure 5. The profile of (left) ozone and (right) potential temperature over Linan from 10–13 March 2001.

troposphere the O_3 mixing ratio ranged from 50 to 75 ppbv. The O_3 profile showed a more complex picture: a fairly sharp increase from the surface to reach a distinct local peak at ~ 3.0 km, with a mixing ratio of 74 ppbv. The O_3 mixing ratio remained at ~ 60 – 70 ppbv up to 10.5 km, from which it showed a steady decreasing trend, reaching a minimum of ~ 50 ppbv at ~ 14.0 km. It then increased sharply at higher altitudes.

[9] We have compared the averaged O_3 profile observed over Hong Kong in 2001 to the monthly average profiles observed from October 1993 to December 2001. We noted that the low O_3 mixing ratio in the upper troposphere over Hong Kong in March and April 2001 was not usually observed in the same months in other years. The O_3 mixing ratio in March and April usually shows a steady increasing trend from the middle to upper troposphere (Figure 3). Instead, the general appearance of the averaged profile in 2001 looked like those observed in the wintertime, for instance, in December and February. One exception is the similarity between the local O_3 peaks over the lower troposphere of Hong Kong and Kunming, which looked quite like that of the averaged enhancement O_3 profile reported by Chan *et al.* [2003b] in the lower troposphere (2.0–6.0 km) from late February to May between 1994 and 1999 over Hong Kong.

3.2. Source and Origin of Ozone in the Upper Troposphere

[10] Chan *et al.* [1998] attributed the occurrence of low O_3 in late autumn and wintertime over Hong Kong's upper troposphere to the injection of O_3 -depleted air masses from the tropical low-level convergence zone near the Indonesian region, following the convective arm of the eastern Asia local Hadley circulation. This kind of transport of O_3 -depleted air masses has been confirmed by the O_3 and trace

gas measurements on board the DC-8 aircraft during the PEM-West B experiment [Newell *et al.*, 1997; Kawakami *et al.*, 1997]. Figure 4 shows the streamlines and isotachs at 150-hPa (~ 14 km) and 200-hPa (~ 12 km) altitudes averaged over 1 March to 13 April 2001. Westerly winds dominated most of Asia and the western Pacific during the TRACE-P period. However, at higher altitudes, especially the 150-hPa and 200-hPa (12 km) altitudes, there was an obvious anticyclonic flow bringing tropical air from the equatorial Pacific near the intertropical convergence zone. This unusual airflow is related to the abnormal convection over tropical SE Asia and Indonesia that occurred during the TRACE-P period [Fuelberg *et al.*, 2003]. The flow influenced most Indo-Chinese regions of SE Asia, the South China Sea, and part of subtropical southeastern China, including Hong Kong and Taiwan. Tropical air has a high moisture content. In fact, the transport of the moisture-rich air had been reflected by a sudden jump in the water mixing ratio profile over Hong Kong at ~ 9.0 km (Figure 2). Judging from the profiles at Kunming and Linan, such a transport process did not affect the tropospheric O_3 over southwestern China and the edge of midlatitude at central eastern China. This is consistent with the flow pattern in Figure 4, which shows that the influence of the tropical air became diminished at higher latitudes, where it was dominated by a westerly flow.

[11] The simultaneous increases in potential temperature and O_3 in the upper troposphere over Linan and the differences in the O_3 profile of this site from that of Hong Kong suggest that there is a different source contributing to the O_3 in the upper troposphere of Linan. Simultaneous increases in O_3 and potential temperature in the troposphere are often considered to be an indication of influences of stratospheric O_3 in the troposphere [Appenzeller and Davies, 1992]. Linan is located at the edge of the middle latitude eastern

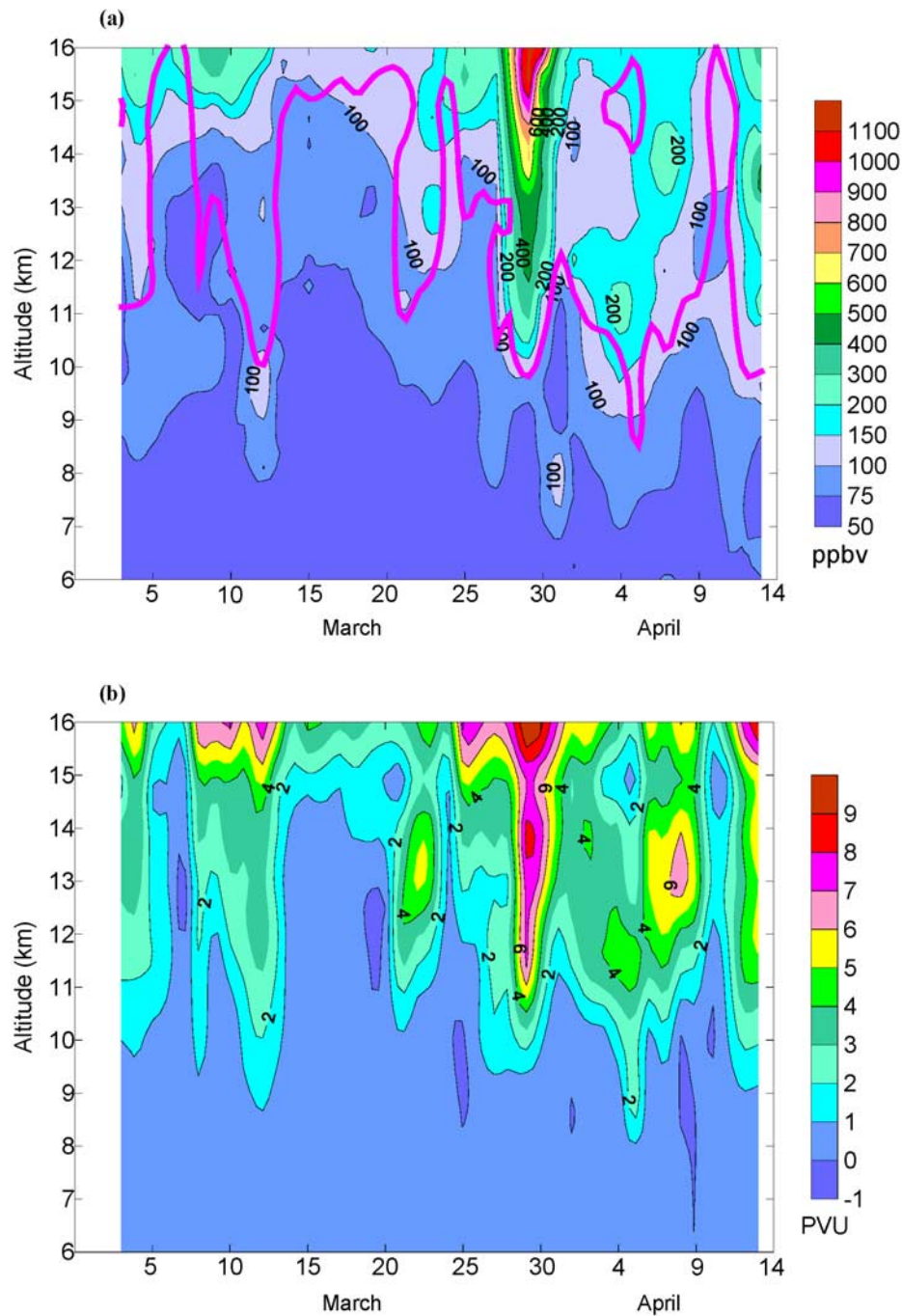


Figure 6. Time-height contour plot of (a) ozone mixing ratios (ppbv) and (b) potential vorticity ($1 \text{ PVU} = 10^{-7} \text{ s}^{-1} \text{ m}^2 \text{ kg}^{-1}$) between 6 and 16 km above sea level over Linan from 3 March to 13 April. The thick line in Figure 6a is the 2-PV unit surface.

Asian coast, where the strongest jet stream across the globe is found in the middle and upper troposphere in springtime [Austin and Midgley, 1994]. This jet stream is a steady feature of winter circulation which intensifies over eastern China and Japan and weakens in the western Pacific. The entrance region of the jet is to the east of 100°E , and the exit region is to the west of 140°E [Ding, 1994]. The jet core of this stream in the TRACE-P period stretched west from over southern Japan to over the central eastern Chinese coast,

including Linan (Figure 4). The wind speed of the jet stream decreased rapidly south of the jet core over Hong Kong and Kunming. The influence of this jet stream over Linan is reflected by the maximum wind speed ($\sim 75 \text{ m s}^{-1}$) at $\sim 12.0 \text{ km}$ altitude (Figure 2). The average maximum wind speed over Hong Kong is $\sim 30 \text{ m s}^{-1}$.

[12] Previous studies in Japan have suggested that the jet stream is very often associated with transport of stratospheric O_3 into the troposphere through tropopause folding and

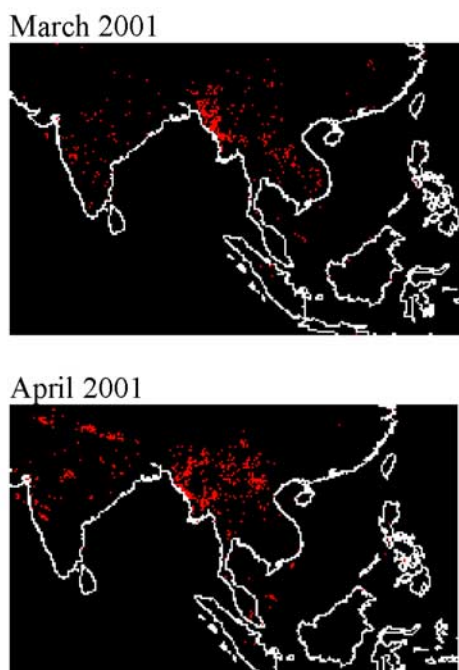


Figure 7. Geographical distributions of fires in Southeast Asia in (top) March 2001 and (bottom) April 2001. The fire locations at a resolution of $1 \times 1 \text{ km}^2$ are denoted by dots.

subsequent diffusion processes [Muramatsu *et al.*, 1984; Wakamatsu *et al.*, 1989; Austin and Midgley, 1994; Tsutsumi and Makino, 1995]. Austin and Midgley [1994] noted that the stratospheric-tropospheric exchange of ozone over northern Japan in spring is associated with the advection of laminae of high and low ozone mixing ratios in the stratosphere. We have frequently observed a similar advection of laminae of high ozone mixing ratios from the lower stratosphere to the upper and middle troposphere over Linan. Such a case is shown by the O_3 profiles in Figure 5 from 10 to 13 March. Within this period, layers of air with high O_3 mixing ratios ($>300 \text{ ppbv}$) advected progressively from the lower stratosphere above 16.0 km to $\sim 15.0 \text{ km}$ on 10 March, 14.8 km (175 ppbv) on 11 March, and 14.8 km (125 ppbv) on 12 March. Note that there were elevated- O_3 layers in the middle and upper troposphere between 8.0 and 15.0 km from 10 to 13 March. These layers were probably the remains of a similar advection of O_3 laminae in earlier periods. The O_3 -rich air in these layers was accompanied by an increase in potential temperature, suggesting that the O_3 is of stratospheric origin (Figure 5).

[13] We have examined the possible relationship between O_3 and potential vorticity (PV) over the middle and upper troposphere of Linan during the experimental period. Figure 6 shows the vertical distribution of the O_3 mixing ratio and PV calculated from National Centers for Environmental Prediction (NCEP) reanalysis data for the whole experimental period. PV is a dynamic tracer of stratospheric air in the troposphere [Danielsen, 1968]. Rather high mixing ratios were observed for O_3 , and there is a prominent vertical variation from the lower stratosphere to the middle troposphere around 8 km asl. Noticeably, regimes of ex-

tremely high O_3 (with a maximum O_3 mixing ratio up to 1200 ppbv) extended from the lower stratosphere deep into the lower altitudes during the beginning, middle, and end of the experiment period. Also, high PV with a value $>10^{-7} \text{ s}^{-1} \text{ m}^2 \text{ kg}^{-1}$ ($1 \text{ PVU} = 10^{-7} \text{ s}^{-1} \text{ m}^2 \text{ kg}^{-1}$) was found at altitudes higher than 8.0 km, with an exception from 6 to 8 March and from 15 to 20 March. The extension of these regimes of high O_3 mixing ratios into the lower altitudes, including the middle and upper troposphere, always corresponded well with the PV. For instance, at altitudes from 12.0 to 14.0 km from 20 March to 7 April, PV values >5 were found at the center of the peak O_3 mixing ratio regions. Note also the high O_3 mixing ratios ($>100 \text{ ppbv}$) below and following the extreme PV regimes (11–12 March and 27 March to 2 April). The high PV values in the middle and upper troposphere over Linan, especially during and after the extremely high O_3 periods, strongly suggest that the O_3 is of stratospheric origin.

[14] The above findings lead us to believe that the high O_3 found in the upper troposphere over Linan in spring 2001 is mainly of stratospheric origin. This is in line with the recent work of Miyazaki *et al.* [2002], who found that most air masses in the upper troposphere over Japan, close to Linan, are not directly affected by the photochemical O_3 formed from emissions in eastern Asia. Since Hong Kong and Kunming are relatively far away from the jet stream, the contributions of stratospheric O_3 to tropospheric O_3 are relatively small. This is supported by the poor relationship between O_3 and PV (not shown).

3.3. Biomass Burning in Southeast Asia as a Source of Ozone in the Lower Troposphere

[15] The occurrence of a similar local O_3 peak in the lower troposphere over Hong Kong and Kunming suggests that there may be a common source leading to such an O_3 maximum. Examination of the individual O_3 profiles from the three launching sites in the year 2001 suggested that elevated O_3 concentrations in the lower troposphere were frequently observed over Hong Kong, less often over Kunming, and scarcely ever over Linan. These O_3 profiles are very similar to those reported by Liu *et al.* [1999] and Chan *et al.* [2000, 2003b]. Chan *et al.* [2000, 2003b] showed that the O_3 enhancements are due to the transport of photochemical O_3 produced from biomass burning emissions from the SE Asian subcontinent.

[16] Chan *et al.* [2003b] used the fire counts deduced from Along Track Scanning Radiometer (ATSR) satellite imagery from the European Space Agency to show that biomass burning activities in the SE Asian subcontinent reached their full strength in the February–April period. In the year 2001, active biomass burning activities also occurred in the SE Asian subcontinent. Using Advanced Very High Resolution Radiometer data, Heald *et al.* [2003] estimated that burning in February–April 2001 was 87% of the climatological average reported by Duncan *et al.* [2003]. These active fire activities were commonly found in the Indo-Burmese region, containing Burma and Laos, and in the Indo-Chinese region, containing northern Thailand. Images from the ATSR World Fire Atlas (<http://shark1.esrin.esa.it/ionia/FIRE/AF/ATSR>) (Figure 7) suggest that such fire activities result in the emission of O_3 precursors such as carbon monoxide (CO), methane, nonmethane hydrocarbons, and

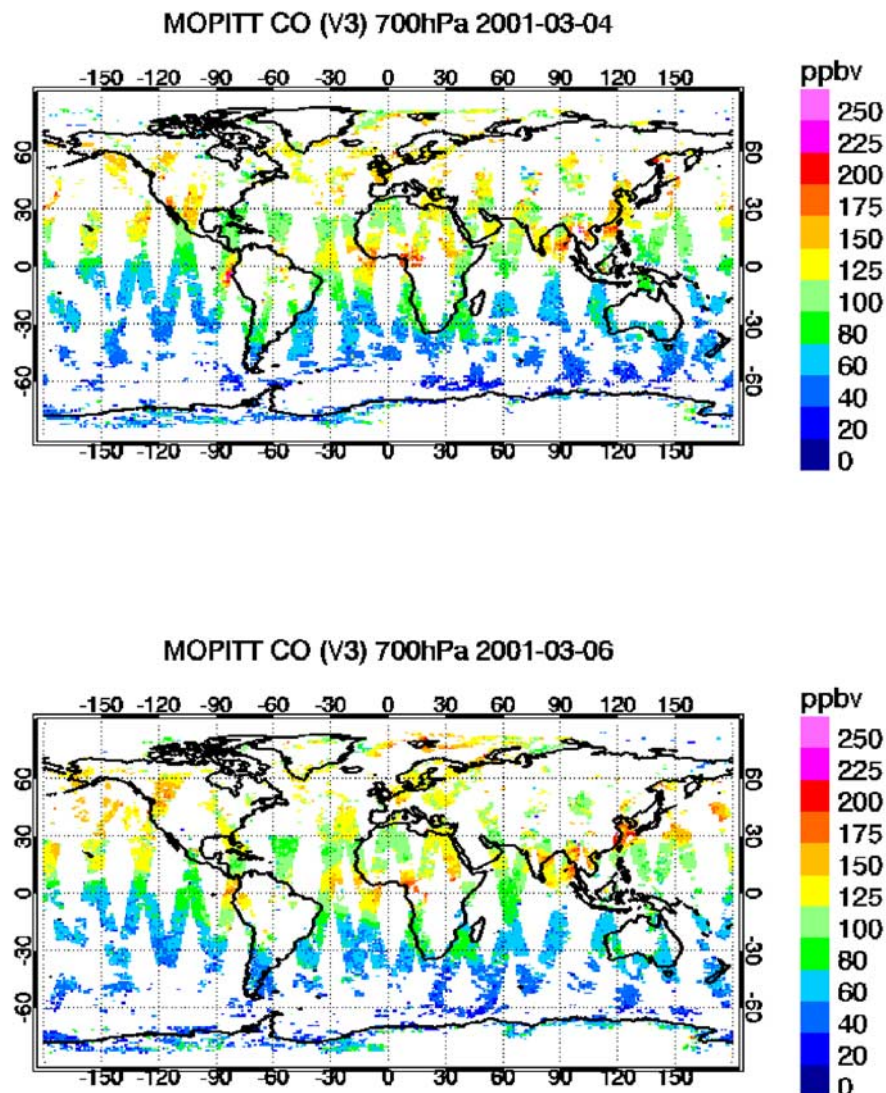


Figure 8. Global distribution of carbon monoxide at 700 hPa altitude on (top) 4 March and (bottom) 6 March 2001.

others into the atmosphere of nearby regions. In fact, enhanced CO columns have been detected by the Measurements of Pollution in the Troposphere (MOPITT) satellite (<http://www.eos.ucar.edu/mopitt/dataimages/index.html>) over the Indo-Burmese and Indo-Chinese regions of SE Asia, close to Yunnan Province, where the Kunming launching site is situated, and downwind over southeastern China, close to Hong Kong (Figure 8). Such high CO levels were confirmed by aircraft measurements along the southeastern Chinese coast [Liu *et al.*, 2003, Figure 5].

[17] Emissions from the burning activities are believed to cause enhancements of tropospheric O_3 in the downwind subtropical Chinese mainland and in the western Pacific. An obvious enhancement case was observed from 21 February to 13 March 2001. Figure 9 shows the O_3 profiles over Hong Kong, Kunming, and Linan within this period. An elevated O_3 layer with a peak concentration at ~ 75 ppbv

started to occur on 21 and 27 February at 2.0–3.8 km altitude over Hong Kong. The peak concentration gained in strength on 2 March and reached full strength, with an O_3 mixing ratio of ~ 105 ppbv, on 7 March, before losing strength on 9 March. Over Kunming a similar enhanced O_3 layer was not observed until 8 March, when O_3 reached a maximum of 100 ppbv at 4.4 km altitude. The enhanced O_3 layer, however, lasted only for a short period of time, until 10 March, when much lower O_3 values were found. The O_3 profiles over Linan showed no noticeable enhancement layer at similar altitudes.

[18] Figure 10 shows the geopotential heights and streamlines for 7 and 8 March. It indicates that there were subtropical anticyclones over the northern Philippines and the South China Sea on 7 and 8 March 2001 which resulted in an anticyclonic flow within the region from 5° – 30° N to 90° – 140° E. Such a flow should have enabled the transport

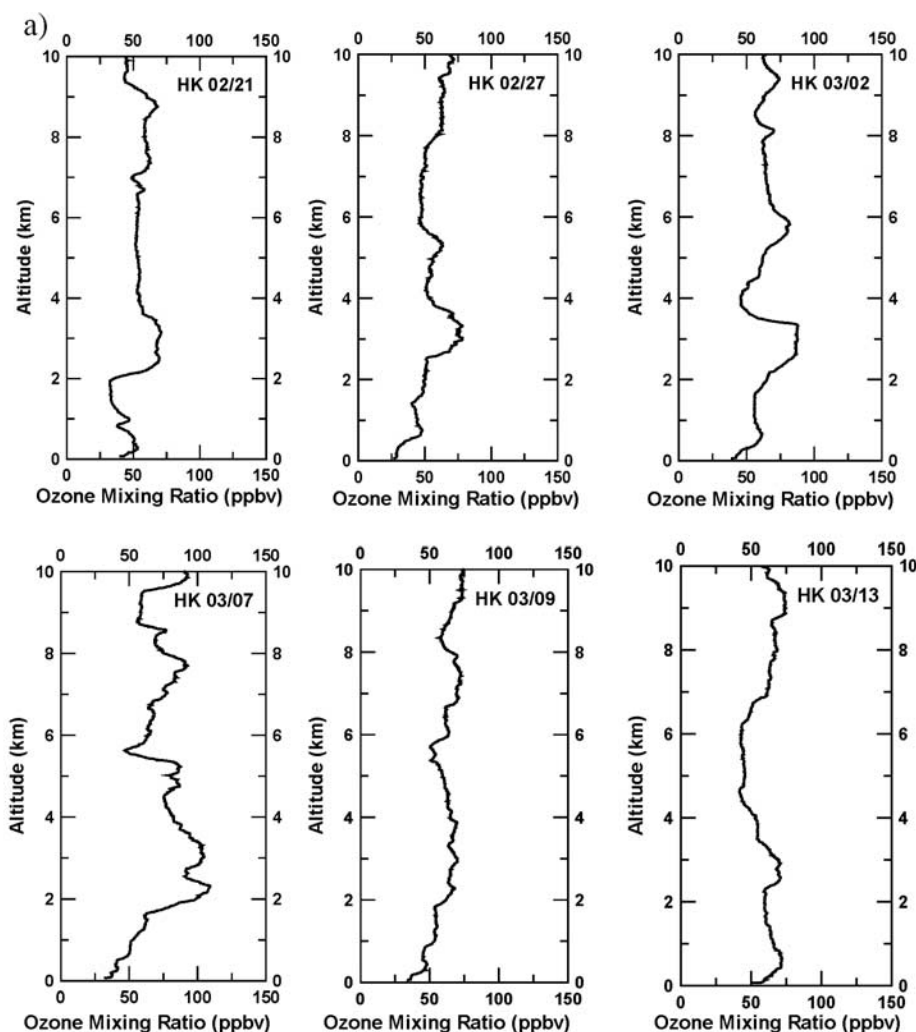


Figure 9. The vertical profiles of ozone over (a) Hong Kong, (b) Kunming, and (c) Linan from 21 February to 13 March 2001.

of biomass burning emissions from the SE Asian subcontinent to the downwind Pacific, including to the nearby vicinity of the SE Asian portion of China, including Kunming and Hong Kong. Figure 11 shows the back air trajectories ending at 3 km over Hong Kong on 7 March and at 4 km over Kunming on 8 March, corresponding to the O_3 peaks. The trajectories were calculated from the meteorological field simulated by Mesoscale Model version 5 using the NCEP reanalysis data as input. The back air trajectories tracked back to the Indo-Burmese and Indo-Chinese regions, where an elevated CO column was observed on 4 March by MOPITT (Figure 8) several days before the air masses reached these sites. We thus believe that the O_3 -enhanced layers over the lower troposphere of Hong Kong and Kunming are similarly caused by biomass burning emissions from the SE Asian subcontinent, as were shown in an earlier study for Hong Kong [Chan *et al.*, 2000, 2003b]. The trajectory at similar altitudes over Linan (not shown), however, showed that the air masses were mainly from the western part of the Chinese mainland. They are believed to be relatively free from the influence of SE Asian biomass burning emissions.

[19] One may consider that industrial activities on mainland China and in eastern Asia emit a lot of O_3 precursors and may contribute to the tropospheric O_3 measured at the three Chinese sites. However, our analysis suggests that stratospheric O_3 is the dominant source of O_3 in the middle and upper troposphere over Linan and that biomass burning in SE Asia causes strong O_3 enhancements in the lower troposphere over Hong Kong and Kunming. It is worthy to point out that episodes with a moderately high O_3 mixing ratio did occur in the boundary layer over Linan, although there was no large O_3 enhancement. Chan *et al.* [2003a] showed that these O_3 episodes are the result of the transport of O_3 and its precursors from the source regions of mainland China.

4. Conclusion

[20] During the TRACE-P period in the spring of 2001 we launched an ozonesonde at three Chinese sites: at subtropical Kunming in southwestern China, close to the SE Asian border, at Hong Kong, on the southeastern Asian coast, and at Linan, at the edge of middle latitudes of central

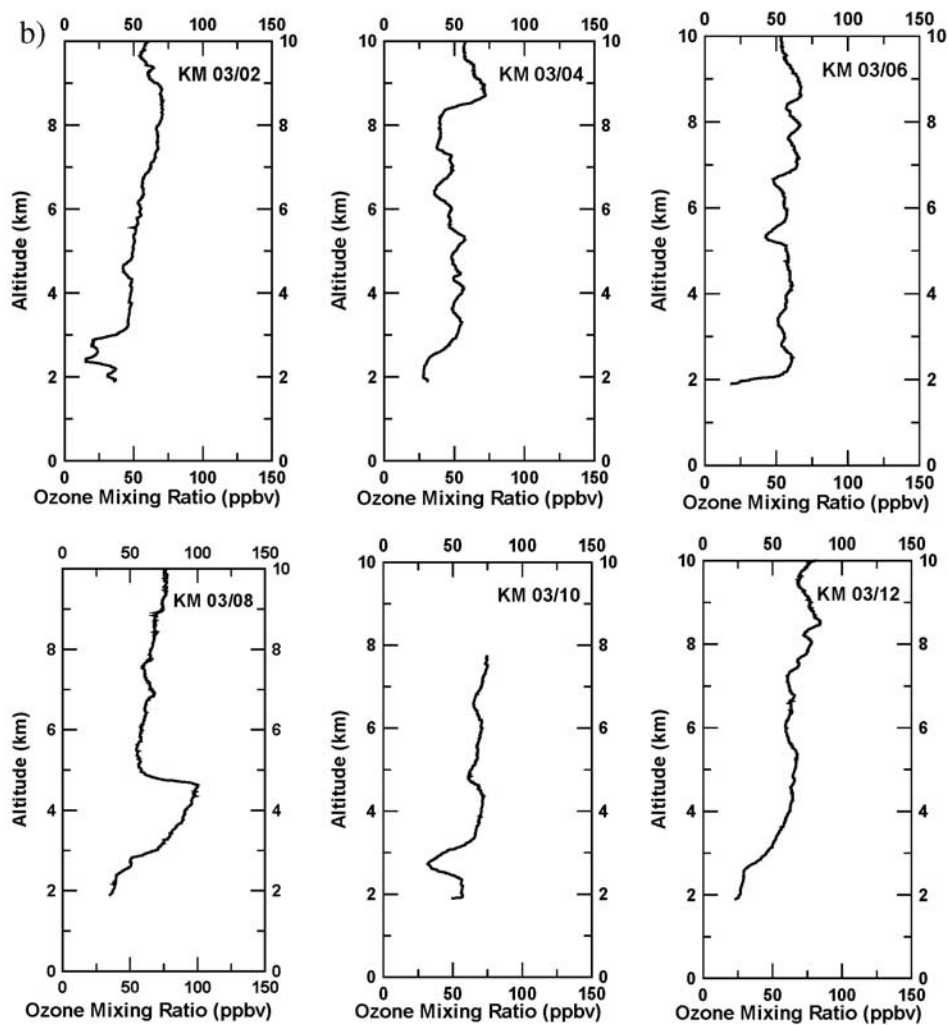


Figure 9. (continued)

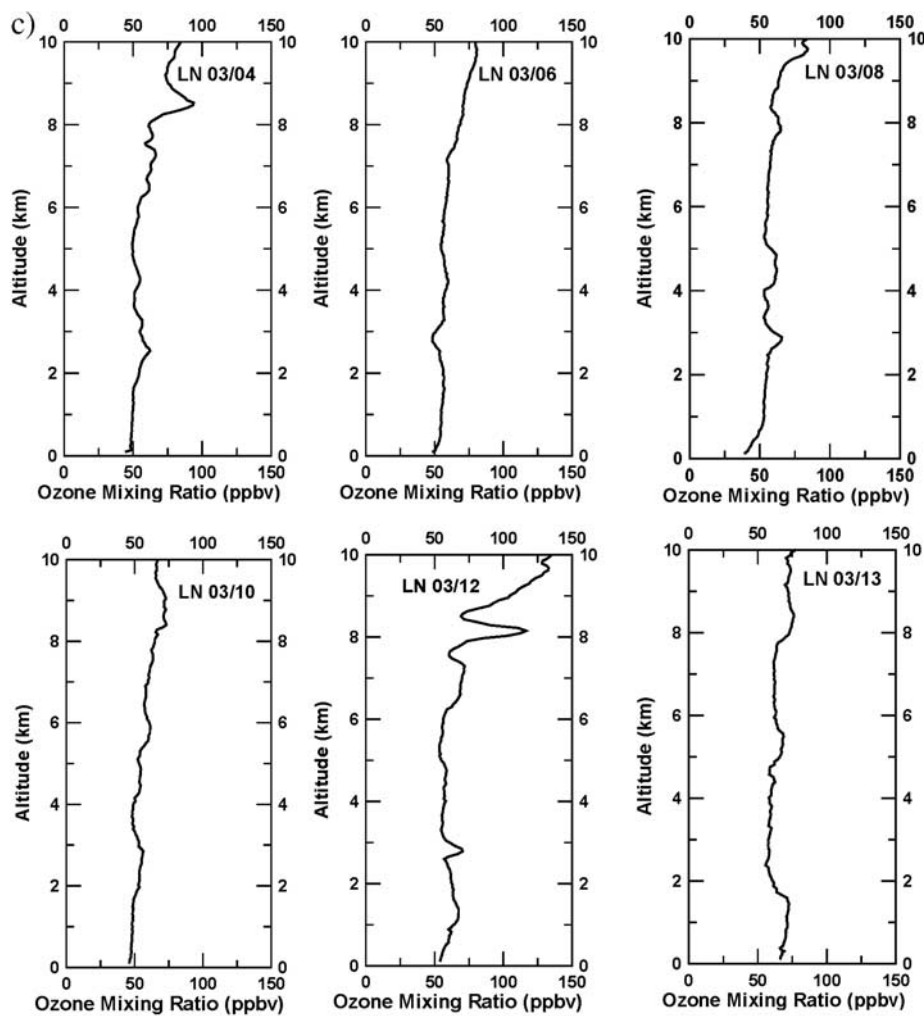
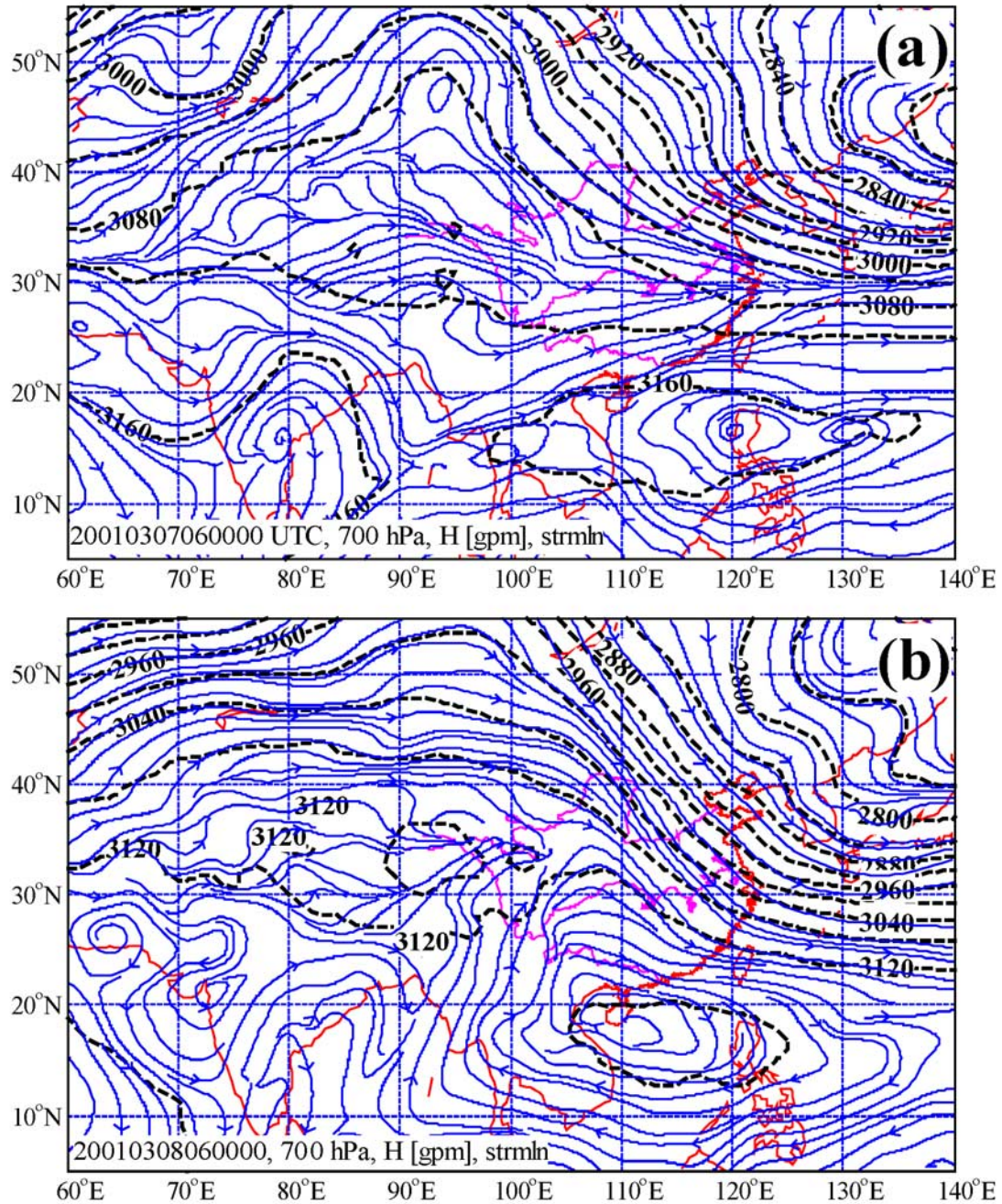


Figure 9. (continued)



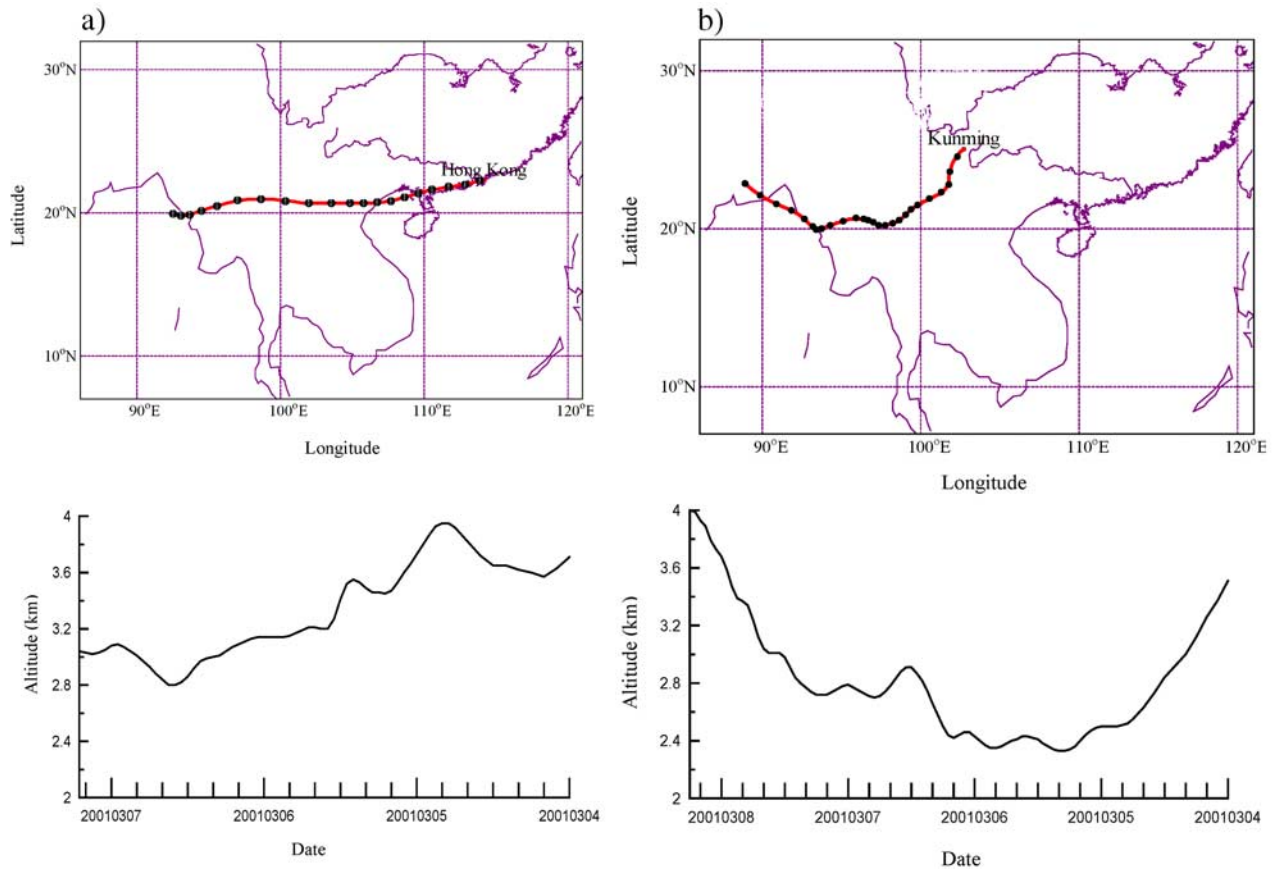


Figure 11. Five-day back air trajectories reaching (a) Hong Kong at 3 km on 7 March 2001 and (b) Kunming at 4 km on 8 March 2001. The labels are in 4-hour intervals.

eastern China. The results show that there are substantial variations in the vertical O_3 distributions over these sites. There were unusually low O_3 values in the upper troposphere of Hong Kong, high O_3 values in the middle and upper troposphere of Linan, and frequent O_3 enhancements in the lower troposphere of Hong Kong and Kunming. The results were analyzed by comparing these ozonesonde data to long-term data from Hong Kong. We also used PV and metrological analyses as well as fire counts and column carbon monoxide information from satellite observations to trace the origins and sources of O_3 .

[21] The low values over the upper troposphere of Hong Kong are the result of the transport of O_3 -depleted air from the intertropical convergence zone of equatorial SE Asian regions following the eastern Asia local Hadley circulation. Such transport processes do not affect the higher latitude of Linan, where stratospheric O_3 is the major contributing source to middle and upper tropospheric O_3 . O_3 enhancements over the lower troposphere of Kunming and Hong Kong are caused by SE Asian biomass burning emissions. Such enhancements are frequently observed over Hong Kong, less often over Kunming, and scarcely ever over Linan, which means that biomass burning emissions from SE Asia in spring 2001 have mainly affected the southern parts of the subtropical Chinese region.

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References

- Appenzeller, C., and H. C. Davies, Structure of stratospheric intrusions into the troposphere, *Nature*, 358, 570–572, 1992.
- Austin, J. F., and R. P. Midgley, The climatology of the jet stream and stratospheric intrusions of ozone over Japan, *Atmos. Environ.*, 28, 39–52, 1994.
- Blake, N. J., D. R. Blake, T.-Y. Chen, J. E. Collins Jr., G. W. Sachse, B. E. Anderson, and F. S. Rowland, Distribution and seasonality of selected hydrocarbons and halocarbons over the western Pacific basin during PEM-West A and PEM-West B, *J. Geophys. Res.*, 102, 28,315–28,333, 1997.
- Chan, C. Y., L. Y. Chan, Y. G. Zheng, J. M. Harris, S. J. Oltmans, and S. Christopher, Effects of the 1997 Indonesian forest fires on tropospheric ozone enhancement, radiative forcing, and temperature change over the Hong Kong region, *J. Geophys. Res.*, 106, 14,875–14,885, 2001.
- Chan, C. Y., L. Y. Chan, H. Cui, X. D. Zheng, Y. G. Zheng, Y. Qin, and Y. S. Li, Origin of the springtime tropospheric ozone maximum over east China at Linan in year 2001, *Tellus, Ser. B*, in press, 2003a.
- Chan, C. Y., L. Y. Chan, J. M. Harris, S. J. Oltmans, D. R. Blake, Y. Qin, Y. G. Zheng, and X. D. Zheng, Characteristics of biomass burning emission sources, transport, and chemical speciation in enhanced springtime tropospheric ozone profile over Hong Kong, *J. Geophys. Res.*, 108(D1), 4015, doi:10.1029/2001JD001555, 2003b.
- Chan, L. Y., H. Y. Liu, K. S. Lam, T. Wang, S. J. Oltmans, and J. M. Harris, Analysis of the seasonal behavior of tropospheric ozone at Hong Kong, *Atmos. Environ.*, 31, 159–168, 1998.
- Chan, L. Y., C. Y. Chan, H. Y. Liu, S. Christopher, S. J. Oltmans, and J. M. Harris, A case study on the biomass burning in Southeast Asia and enhancement of tropospheric ozone over Hong Kong, *Geophys. Res. Lett.*, 27(10), 1479–1483, 2000.

- Danielsen, E. F., Stratosphere-troposphere exchange based on radioactivity, ozone and potential vorticity, *J. Atmos. Sci.*, 25, 502–518, 1968.
- Ding, Y., *Monsoons Over China*, Kluwer Acad., Norwell, Mass., 1994.
- Duncan, B. N., R. V. Martin, A. Staudt, R. Yevich, and J. A. Logan, Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, *J. Geophys. Res.*, 108(D2), 4100, doi:10.1029/2002JD002378, 2003.
- Folkens, I., R. Chatfield, D. Baumgardner, and D. Proffitt, Biomass burning and deep convection in southeastern Asia: Results from ASHOC/MAESA, *J. Geophys. Res.*, 102, 13,291–13,299, 1997.
- Fuelberg, H. E., C. Kiley, J. R. Hannan, D. J. Westberg, M. A. Avery, and R. E. Newell, Meteorological conditions and transport pathways during the transport and chemical evolution over the Pacific (TRACE-P) experiment, *J. Geophys. Res.*, 108(D20), 8782, doi:10.1029/2002JD003092, in press, 2003.
- Fujiwara, M., K. Kita, S. Kawakami, T. Ogawa, N. Komala, S. Saraspriya, and A. Suropto, Tropospheric ozone enhancements during the Indonesian forest fire events in 1994 and in 1997 as revealed by ground-based observations, *Geophys. Res. Lett.*, 26(16), 2417–2420, 1999.
- Harris, J. M., S. J. Oltmans, E. J. Dlugokencky, P. C. Novelli, B. J. Johnson, and T. Mefford, An investigation into the source of the springtime tropospheric ozone maximum at Mauna Loa Observatory, *Geophys. Res. Lett.*, 25(11), 1895–1898, 1998.
- Heald, C., D. J. Jacob, P. I. Palmer, M. J. Evans, G. W. Sachse, H. Singh, and D. Blake, Biomass burning emission inventory with daily resolution: Application to aircraft observations of Asian outflow, *J. Geophys. Res.*, 108(D21), 8811, doi:10.1029/2002JD003082, in press, 2003.
- Jacob, D. J., J. Crawford, M. M. Kleb, V. S. Connors, R. J. Bendura, J. L. Raper, G. W. Sachse, J. Gille, L. Emmons, and J. C. Heald, Transport and chemical evolution over the Pacific (TRACE-P) mission: Design, execution, and first results, *J. Geophys. Res.*, 108(D20), 8781, doi:10.1029/2002JD003276, in press, 2003.
- Kawakami, K., et al., Impact of lightning and convection on reactive nitrogen in the tropical free troposphere, *J. Geophys. Res.*, 102, 28,367–28,384, 1997.
- Komhyr, W. D., R. A. Barnes, G. B. Brothers, J. A. Lathrop, and D. P. Opperman, Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, *J. Geophys. Res.*, 100, 9231–9244, 1995.
- Lelieveld, J., et al., The Indian Ocean Experiment: Widespread air pollution from south and Southeast Asia, *Science*, 291, 1031–1036, 2001.
- Liu, H. Y., W. L. Chang, S. J. Oltmans, L. Y. Chan, and J. M. Harris, On springtime high ozone events in the lower troposphere from SE Asian biomass burning, *Atmos. Environ.*, 33, 2403–2410, 1999.
- Liu, H., D. J. Jacob, I. Bey, R. Yantosca, B. N. Duncan, and G. W. Sachse, Transport pathways for Asian pollution outflow over the Pacific: Interannual and seasonal variations, *J. Geophys. Res.*, 108(D20), 8786, doi:10.1029/2002JD003102, in press, 2003.
- Matsueda, H., and H. Y. Inoue, Aircraft measurements of trace gases between Japan and Singapore in October of 1993, 1996, and 1997, *Geophys. Res. Lett.*, 26(16), 2413–2416, 1999.
- Miyazaki, Y., K. Kita, Y. Kondo, M. Koike, M. Ko, W. Hu, S. Kawakami, D. R. Blake, and T. Ogawa, Springtime photochemical ozone production observed in the upper troposphere over east Asia, *J. Geophys. Res.*, 108(D3), 8398, doi:10.1029/2001JD000811, 2002.
- Muramatsu, H., T. Sasaki, M. Hirota, and Y. Makino, An aircraft observation of an intrusion process of stratospheric ozone into the troposphere, *Meteorol. Geophys.*, 35, 1–10, 1984.
- Newell, R. E., E. V. Browell, D. D. Davis, and S. C. Liu, Western Pacific tropospheric ozone and potential vorticity: Implications for Asian pollution, *Geophys. Res. Lett.*, 24, 2733–2736, 1997.
- Pochanart, P., J. Kreasuwun, P. Sukasem, W. Geerathadaniyom, M. S. Tabucanon, J. Hirokawa, Y. Kajii, and H. Akimoto, Tropical tropospheric ozone observed in Thailand, *Atmos. Environ.*, 35, 2657, 2001.
- Talbot, R. W., et al., Chemical characteristics of continental outflow from Asia to the troposphere over the western Pacific Ocean during September–October 1991: Results from PEM-West A, *J. Geophys. Res.*, 101, 1713–1726, 1996.
- Thompson, A. M., J. C. Witte, R. D. Hudson, H. Guo, J. R. Herman, and M. Fujiwara, Tropical tropospheric ozone and biomass burning, *Science*, 291, 2128–2132, 2001.
- Tsutsumi, Y., and Y. Makino, Vertical distribution of the tropospheric ozone over Japan: The origin of the ozone peaks, *J. Meteorol. Soc. Jpn.*, 73, 1041–1058, 1995.
- Tsutsumi, Y., Y. Sawa, Y. Makino, J. B. Jensen, J. L. Gras, B. F. Ryan, S. Diharto, and H. Harjanto, Aircraft measurements of ozone, NO_x, CO, and aerosol concentrations in biomass burning smoke over Indonesia and Australia in October 1997: Depleted ozone layer at lower altitude over Indonesia, *Geophys. Res. Lett.*, 26(5), 595–598, 1999.
- United Nations Environment Programme and Centre for Clouds, Chemistry and Climate, *The Asian Brown Cloud: Climate and Other Environmental Impacts*, U.N. Environ. Prog., Nairobi, 2002.
- Wakamatsu, S., I. Uno, H. Ueda, K. Uehara, and H. Tateishi, Observational study of stratospheric ozone intrusions into the lower troposphere, *Atmos. Environ.*, 23, 1815–1826, 1989.
- Yonemura, S., H. Tsuruta, S. Kawashima, S. Sudo, L. C. Peng, L. S. Fook, Z. Johar, and M. Hayashi, Tropospheric ozone climatology over Peninsular Malaysia from 1992 to 1999, *J. Geophys. Res.*, 107(D15), 4229, doi:10.1029/2001JD000993, 2002a.
- Yonemura, S., H. Tsuruta, T. Maeda, S. Kawashima, S. Sudo, and M. Hayashi, Tropospheric ozone variability over Singapore from August 1996 to December 1999, *Atmos. Environ.*, 36, 261–270, 2002b.

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